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SUPERCONDUCTORS, MAGNETIC DETECTORS AND MAGNETIC ELECTRONICS.(U)  
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DIVISION OF ENGINEERING AND APPLIED PHYSICS

GORDON MCKAY LABORATORY

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FINAL REPORT

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M. Tinkham, Principal Investigator

Superconductors, Magnetic Detectors and Magnetic Electronics.

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Introduction

With the termination of this contract on Sept. 30, 1976, continuous ONR support extending over 18 years, including 8 years at the University of California, Berkeley, has been interrupted. New ONR support has been requested under a different program, and it is hoped that the interruption of support will be brief. None the less, it is necessary and appropriate to review at this time the accomplishments made with the assistance of this ONR support. (There has also been continuous NSF support during this period, and occasional support from other agencies, but because of the healthy mutual interaction of the various projects, it is not really feasible to assign unique support to each piece of work.)

A final report on the Berkeley work was submitted on July 15, 1966. This report covers our pioneering work in opening up the far infrared to useful application in the study of solids: finding and studying the energy gap in superconductors, first observation of antiferromagnetic resonance and exchange resonances in the far infrared, first observation of the temperature-dependent soft mode in ferroelectric crystals, first observation of the far-infrared absorption due to random charge in disordered systems, etc. This report also outlined the early phases of our work on the effect of magnetic fields in creating gapless superconductors and on the implications of fluxoid quantization for the critical fields of thin films and bulk superconductors. Some 41 journal articles and other publications as well as 14 Ph.D. theses resulted from this work.

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The present report is intended to provide a similar outline of the work accomplished at Harvard in the years 1966-76. This work has been reported in the 12 Technical Reports and 49 other publications listed later in this Report, and could not possibly be summarized completely here. Instead, I shall give only a rather cursory survey, drawing attention to some of the specific advances reported in these publications.

### Brief Summary of Research Carried Out

Far Infrared: Our early Harvard work in the far infrared ( $\lambda = 0.1 - 1 \text{ mm}$ ) was carried out using a mercury arc source and a grating monochromator built in the Gordon McKay Laboratory machine shop. With the use of digital recording and reduction of data, this technique was adequate for the pioneering work of Torrance on the complex multi-magnon excitation spectrum of  $\text{CoCl}_2 \cdot 2\text{H}_2\text{O}$  ("CC2"). This material, whose magnetic order consists of linear chains of highly anisotropic Co ions, changes from an antiferromagnetic to a ferrimagnetic to a ferromagnetic phase as the applied magnetic field is increased. In all of these phases, it was possible to observe transitions in which many spins flipped at once, leading to enormous effective g-values.

This initial work was followed by that of Nicoli, who designed and built an HCN/DCN laser source which supplied strong monochromatic radiation at 337 and 311 microns. Used with a  $\text{Nb}_3\text{Sn}$  superconducting magnet capable of reaching 125 kG, this source allowed the spectrum of CC2 to be re-examined at high resolution. This reexamination allowed us to observe transitions in which as many as 15 spins flipped at once, and allowed a successful quantitative test of our theory of line intensities as well as line positions.

The same laser and magnet combination was used by Peech to carry out a high-resolution study of the cyclotron resonance of copper. Because of the higher frequency of the far infrared, distinct mass variations over the Fermi surface could be resolved which did not show up in previous microwave studies.

These applications of the HCN laser demonstrated how much could be gained by replacing the weak broad-band monochromator source by a more intense monochromatic laser source, but it also demonstrated the limitations

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imposed by having only a few spot frequencies to work with. To rectify this difficulty, Weitz has built a more versatile far-infrared gas laser, optically pumped by a 30 watt (CW)  $\text{CO}_2$  laser. The initial configuration used a large optical cavity defined by 4" mirrors in a pyrex tube. Although this lased on many lines of  $\text{CH}_3\text{OH}$  and  $\text{CH}_3\text{F}$  in the range 119-570  $\mu\text{m}$ , and was used successfully in preliminary measurements, the output mode structure was not well adapted to focussing on a small detector because of the coupling holes in the centers of the mirrors. This difficulty was cured by rebuilding the laser in the waveguide configuration, coupling the output uniformly from one end through a partially transmitting end mirror. The resulting beam is near to the ideal gaussian form, and the efficiency is also higher, giving rise to stronger output. Initial experiments have been carried out, in which this single-mode far-infrared beam is used to irradiate superconducting point contact detectors, inducing steps in the I-V characteristic at the voltage  $V_n = n\hbar\omega/2e$  expected from the Josephson relation. These experiments have shown the directional and polarization dependence expected from simple long-wire antenna theory.

Superconductivity: Our early work in this field (while still in Berkeley) emphasized the energy gap aspects of superconductors: the first far-infrared measurements of the width of the gap in many materials; quantitative measurements of the complex conductivity of superconducting films near the gap frequency, culminating in the discovery of the strong-coupling corrections in lead; measurements of the tunneling density of states and other properties in films in the presence of magnetic fields, supporting the theory of the approach to gapless superconductivity. These studies of the effects of magnetic fields led to a paper pointing out the important implications of fluxoid quantization in superconductors, which in turn led to a simple derivation of the upper critical field  $H_{c2}$  of type II superconductors, a theory of the angular dependence of the critical field of thin films, and the interpretation of the flux-modulated  $T_c$  of superconducting cylinders observed by Little and Parks. The work also led to the Harvard experiments of Goren, who showed the existence of an array of flux spots in thin films in weak normal fields.

The next major step in the evolution of our program occurred when

Dr. M. R. Beasley joined the group in Harvard, bringing his expertise in superconducting quantum devices (SQUID's) based on the Josephson effect. Our first application of these devices was Collub's use of a SQUID magnetometer to make quantitative measurements of the tiny (six orders of magnitude below the Meissner effect perfect diamagnetism) enhanced diamagnetism due to thermodynamic fluctuations in superconductors up to temperatures of twice  $T_c$ . Although his results were in qualitative agreement with Ginzburg-Landau theory, subsequent theoretical work showed that major quantitative discrepancies could be traced to the importance of short-wavelength modes in the fluctuation spectrum. Tai used the same basic technique to make precise measurements of the anisotropy in the penetration depth of magnetic fields below  $T_c$  in single crystal superconductors. Later, Prober applied this approach to the superconducting layered compounds  $TaS_2$  and  $NbSe_2$ , with and without intercalation by organic compounds, finding 3-dimensional behavior despite the extreme anisotropy. In a different sort of application, Newbower used a SQUID in the SLUG (Superconducting Low-inductance Undulating Galvanometer) configuration to measure the onset of resistance in filamentary superconductors in the form of micron-diameter tin whiskers. This work (together with similar work of Webb's group at Cornell) gave a decisive verification of the Langer-Ambegaokar-McCumber-Halperin theory of resistance in superconductors by thermally activated phase slip processes. These are the processes which determine the (astrophysical) time scale for decay of persistent currents in superconductors.

In the course of this work (and of the similar work at Cornell), it was noticed that with larger currents the resistance of the whisker did not appear smoothly in a narrow range of temperature, but in a series of well-defined steps extending over a considerable range of temperature (or current). The origin of these steps remained a mystery for some years, but it was uncovered by the thesis work of W. J. Skocpol, who continued in the group as an Assistant Professor of Physics. By studying the I-V characteristics of superconducting microbridges of various configurations, Skocpol found a clear explanation of the superiority of point-contact weak links: the essentially three-dimensional geometry allows the most efficient possible cooling to prevent temperature rise due to Joule heating in the finite voltage regime. By working near enough to  $T_c$ , where critical currents



are small, these heating effects can be made negligible, and one reaches the regime dominated by the "steps" in the I-V curve. Skocpol's work showed that each step corresponds to the creation of another quantum-phase-slip center, localized at some point along the length of the microbridge. The ac supercurrents in these centers occur at frequencies  $f$  given by the Josephson relation  $f = 2eV/h$ , where  $V$  is the voltage drop across a single center, not the entire bridge, and  $h$  is Planck's constant. A key quantitative discovery from our analysis is that the dynamic resistance of these steps is determined by the inelastic scattering time  $\tau_2$  of the electrons due to phonons. This, in turn, implies that the limiting relaxation time is not the Ginzburg-Landau time  $\tau_{GL}$  as had previously been supposed in all theoretical work on time-dependent superconductivity, but this essentially normal-electron relaxation time.

This discovery has launched us into a continuing study of the nature of the non-equilibrium in time-dependent superconductivity, a topic which has obvious importance for understanding what limits the potential high-frequency performance of superconducting devices. This work is now proceeding rapidly on several fronts. Octavio has demonstrated the expected superiority of variable thickness tin microbridges (i.e. - thick "banks", thin bridge) by observing 186 steps induced by X-band microwaves, corresponding to Josephson effect operation at up to  $V = 3.7$  mV. This is a higher voltage than had ever been reported for a microbridge of any material. We have developed a quantitative theory of the limitations to finite voltage operation set by heating even in the favorable 3-dimensional cooling geometry, which appears to account very well for the best performance achieved to date in variable thickness bridges and in point contacts. Further experimental work is underway to probe the nature of the disequilibrium created by irradiation by lasers (both far IR and visible), as well as to probe the effect of external pairbreakers (such as magnetic fields) on the operation of the phase-slip centers in bridges operating at finite voltage. Unfortunately, only preliminary results from these experiments are available at this time, and publication will require further work.

Brief mention should also be made of the work of Davidson in developing a high-performance ultra-low-noise amplifier, using a SQUID as the sensitive element. He showed that such current-controlled amplifiers are mathematical

"duals" of the high-impedance voltage-controlled FET amplifiers, and hence they offer complementary performance optimized for low-impedance applications. Davidson also applied this sensitive voltmeter to detection of resistance in the microcomposite "Tsuei wire" which contains metallurgically produced superconductive filaments of finite length in a normal metal matrix, and should show a resistance about  $10^8$  times lower than pure copper even if the copper is fully normal. Only a superconductive amplifier can distinguish such small resistances from perfect conductivity in a short sample test.

Finally, we mention the work of Powell on novel ways of preparing superconductive sintered compacts of NbN and NbC microcrystallites. Although his samples were somewhat superior to those produced by other techniques, they do not appear technologically useful relative to other materials already available.

List of Technical Reports

|   | <u>Date</u> | <u>Title</u>  |
|---|-------------|---|
| 1. J.B. Torrance, Jr.                           | 1968        | Excitation of Multiple Magnon Bound States in Anisotropic Linear Chains   |
| 2. R.N. Goren                                   | 1969        | Patterns of Magnetic Flux Penetration in Superconducting Films  |
| 3. J.P. Gollub                                  | 1970        | Diamagnetism due to Fluctuations in Superconductors   |
| 4. R.S. Newbower                                | 1971        | Femtovolt Measurements of the Superconducting Transition in Tin Whisker Crystals                                      |
| 5. D.F. Nicoli                                  | 1972        | Far Infrared Laser Spectroscopy of Spin-Clusters in the Linear Ising System $\text{CoCl}_2 \cdot 2\text{H}_2\text{O}$ |
| 6. P.C.L. Tai                                   | 1973        | The Anisotropy of the Penetration Depth in Superconducting Tin  |
| 7. A. Davidson<br>R.S. Newbower<br>M.R. Beasley | 1974        | An Ultra-Low-Noise Preamplifier Using Superconducting Quantum Devices   |
| 8. W.J. Skocpol                                 | 1974        | Electrical Behavior of Superconducting Microbridges   |
| 9. J.M. Peech                                   | 1974        | Far Infrared Cyclotron Resonance in Metals  |
| 10. D.E. Prober                                 | 1975        | Magnetic Properties of Superconducting Layered Compounds  |
| 11. R.M. Powell                                 | 1975        | Preparation and Superconducting Properties of Sintered Compacts of NbN Microcrystallites                              |
| 12. A. Davidson                                 | 1975        | Applied Superconductivity: Amplifiers and Microcomposite Wire   |



### List of Publications

1. Angular Dependence of the Pinning and the Flow of Vortices in Thin Superconducting Films; R. Deltour and M. Tinkham, *Physics Letters* 23, 183 (1966).
2. An Experimentalist's View of Superconductivity, *Quantum Fluids*; M. Tinkham, Edited by D.F. Brewer, North-Holland Press, 1966, p. 353.
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12. Magnon-Phonon Interaction Observed in Far Infrared Studies of  $\text{FeCl}_2 \cdot 2\text{H}_2\text{O}$ ; K.A. Hay and J.B. Torrance, Jr., *J. Appl. Phys.* 40, 999 (1969).
13. Superconductivity; M. Tinkham, invited paper, New York Meeting of Am. Phys. Soc., February, 1969.
14. Observation of Enhanced Diamagnetism above  $T_c$  in Indium due to Thermodynamic Fluctuations; J.P. Gollub, M.R. Beasley, R.S. Newbower, and M. Tinkham, *Phys. Rev. Letters* 22, 1288 (1969).
15. Magnon Bound States in Anisotropic Linear Chains; J.B. Torrance, Jr., and M. Tinkham, *Phys. Rev.* 187, 587 (1969).
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20. Fluctuation Effects in the Magnetic Transitions of Bulk Superconductors; J.P. Gollub, M.R. Beasley, and M. Tinkham, Proceeding 12th Int. Conf. on Low Temp. Phys., Kyoto, 1970; Edited by E. Kanda, Keigaku Publ. Co., Tokyo, p. 271.
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48. Preparation and Superconducting Properties of Ultrafine Powders and Sintered Compacts of NbC and NbN; R.M. Powell, W.J. Skocpol, and M. Tinkham, J. Appl. Phys., to appear.
49. Heating Effects in High-Frequency Metallic Josephson Devices: Voltage Limit, Bolometric Mixing, and Noise; M. Tinkham, M. Octavio, and W.J. Skocpol, J. Appl. Phys., to appear.